



11 Publication number:

0 533 160 A1

(12)

## **EUROPEAN PATENT APPLICATION**

(21) Application number: 92115931.5

(51) Int. Cl.5: C08L 23/04

2 Date of filing: 17.09.92

3 Priority: 18.09.91 US 761603

Date of publication of application:24.03.93 Bulletin 93/12

Designated Contracting States:
 BE DE ES GB

7) Applicant: PHILLIPS PETROLEUM COMPANY 5th and Keeler Bartlesville Oklahoma 74004(US)

2 Inventor: Coutant, William Richard 5840 Meadowcrest Bartlesville, OK 74006(US) Inventor: Martin, Joel Leonard 636 SE Kenwood Drive Bartlesville, OK 74006(US)

Papersentative: Geissler, Bernhard, Dr.jur.,
Dipl.-Phys. et al
Patent- u. Rechtsanwälte Bardehle
-Pagenberg-Dost-Altenburg
-Frohwitter-Geissler & Partner Postfach 86 06
20 Galileiplatz 1
W-8000 München 80 (DE)

Polyethylene blends.

② A composition of matter comprising: (a) 10 to 80 weight percent inclusive of a low molecular weight polyethylene resin having a density of at least 0.94 grams per cubic centimeter, a melt index greater than 25 grams per 10 minutes, and a heterogeneity index between 2 and 12 inclusive; and (b) 20 to 90 weight percent inclusive of a high molecular weight ethylene copolymer having a density less than 0.955 grams per cubic centimeter, a high load melt index between 2 and 10 grams per 10 minutes inclusive, and a heterogeneity index between 2 and 10 inclusive; wherein said composition of matter has a melt index greater than 0.05 grams per 10 minutes.

### **BACKGROUND OF THE INVENTION**

10

15

50

This invention relates to polyethylene blends.

Colorless polymeric materials range from being almost totally transparent to being opaque. The opacity is related to the light scattering process occurring within, and at the surface of, the material. Incidental radiation passes through non-absorbing, isotropic, and optically homogenous samples with essentially little loss in radiation intensity. Actually, all polymeric materials scattered light to some extent. The angular distribution of the scattered light is a complex phenomenon because of the scattering due to micromolecular interactions.

Transparency is defined, in general, as a state permitting perception of objects through a sample. In more specific terms, transparency is the amount of undeviated light, that is, the original intensity minus all light absorbed, scattered, or lost through any other means. This scattered light reduces the contrast between light, dark, and other colored parts of objects viewed through the material and produces a milkiness or haze in the transmitted image.

Haze is a measure of the amount of light deviating from the direction of transmittancy of the light by et least 2.5 degrees. In more specific terms, haze is that percentage of transmitted light that in passing through the specimen deviates from the incident beam by forward scattering. In commercial hazemeters, only light deviating more than about 2.5 degrees from the transmitted beam direction is considered haze. The effect of this haze is to impart a cloudy or milky appearance to a sample.

Commercial products are desired that have low haze values. The reasons for wanting such low haze products vary from purely safety purposes to purely marketing purposes. Consequently, methods to produce materials which have low haze values are of great scientific and economic value.

Polyethylene is known as a translucent to opaque white polymer with the opacity increasing with the density. One of the by-products of this is that the haze tends to increase in polyethylene as the density increases. That is, without optically modifying polyethylene, a polyethylene sample which has a density of 0.93 should have a haze value less than a polyethylene sample which has a density of 0.97 when made under similar conditions. However, in many applications it is desirable to use an ethylene polymer which has a high density because of the desirable properties of high density polyethylene. For example, tensile strength, hardness, chemical resistance, and flexural modulus all tend to increase with increasing density. Consequently, providing a polyethylene resin with a low haze value and a high density would be both scientifically and economically valuable.

It is known in the art to blend components with dissimilar characteristics to achieve a blend with superior performance properties. However, this task is not easy to accomplish. For example, the components of a blend can interact antagonistically resulting in a lowering of the expected performance of the blend, or the components can interact producing a blend that is only the mere aggregation of the separate component parts.

Thermoplastic blends are very desirable for applications such as pipes, films, and bottles. However, each application requires a resin tailored for that particular application. Furthermore, each of these tailored resins are judged by many performance attributes. While it is possible to improve one attribute it is often necessary to find a compromise of the various attributes in order to find a resin which fulfills the customer's requirements.

This invention provides an improved compromise of characteristics thereby increasing the availability of possible solutions to consumer demands.

## 5 SUMMARY OF THE INVENTION

It is an object of this invention to provide an improved polyethylene blend.

It is another object of this invention to provide a polyethylene blend which has improved optical properties.

It is still another object of this invention to provide a polyethylene blend with improved physical properties.

These and other objects of this invention will become apparent to those skilled in the art from the following detailed description of the invention.

In accordance with this invention, a composition of matter is provided comprising: (a) 10 to 80 weight percent inclusive of a low molecular weight polyethylene resin having a density of at least 0.94 grams per cubic centimeter, a melt index greater than 25 grams per 10 minutes, and a heterogeneity index between 2 and 12 inclusive; and (b) 20 to 90 weight percent inclusive of a high molecular weight ethylene copolymer having a density less than 0.955 grams per cubic centimeter, a high load melt index between 2 and 10

grams per 10 minutes inclusive, and a heterogeneity index between 2 and 10 inclusive; wherein said composition of matter has a melt index greater than 0.05 grams per 10 minutes.

### **DETAILED DESCRIPTION OF THE INVENTION**

The composition of this invention comprises two components. The first component is a low molecular weight polyethylene resin. The second component is a high molecular weight polyethylene resin. Further information concerning the characteristics of these two components are given below.

Polyethylene is a thermoplastic material available in a wide range of flow rates and densities. Polyethylene offers useful properties such as toughness at a variety of temperatures, stiffness ranging from flexible to rigid, as well as excellent chemical resistance. Currently, polyethylene can be fabricated and processed by most known thermoplastic processes in the art.

Polyethylene is generally classified primarily on the basis of two characteristics, namely, density and flow rate (commonly identified as melt index). ASTMD 1248-84 (reapproved 1989) provides a convenient system for categorizing polyethylenes. First, polyethylenes are generally classified according to density: type I polyethylene has a nominal density between 0.910-0.925 grams/cubic centimeter (g/cm³); type II polyethylene has a nominal density between 0.926-0.940 g/cm³; type III polyethylene has a nominal density between 0.941-0.959 g/cm³; and type IV polyethylene has a nominal density of 0.960 g/cm³ or above. Type I polyethylene is usually defined as a low density resin. Type II polyethylene is usually classified as a medium density resin. Type III and IV polyethylenes are usually classified as high density resins.

Within each density type there exists five different categories of flow rates. A category 1 polyethylene has a nominal flow rate greater than 25 g/10 min. at 190°C and a 2160 gram load. A category 2 polyethylene has a nominal flow rate greater than 10 and up to and including 25 g/10min. A category 3 polyethylene has a nominal flow rate greater than 1 up to and including 10 g/10min. A category 4 polyethylene has a nominal flow rate greater than 0.4 up to and including 1.0 g/10min. A category 5 polyethylene has a nominal flow rate equal to or less than 0.4 g/10min.

These types and categories of polyethylene can be made by various processes known in the art. For example, types I and II polyethylene can be made by a high pressure process which uses oxygen, peroxide and/or other strong oxidizers as a catalyst. Pressures of reaction ranging from 100 to 350 MPa are typically employed. Polyethylene resins formed in these types of processes are highly branched with short side chains occurring every 15 to 40 carbon atoms on the chain backbone. Furthermore, the crystallinity of these types of polyethylene resins is approximately 40 to 60 percent with the amorphous content of the polymer increasing as the density is reduced.

On the other hand, types I, II, III, and IV polyethylene can be produced from such processes as slurry, solution, or gas phase processes. These processes are generally known as low pressure processes. Catalysts used in these processes vary widely, but the most frequently used are transition metals in combination with metal halides or activated metal oxides. Reaction pressures normally fall within 50 to 800 psi. The polyethylene produced by these processes is more linear in nature, that is, little or no long chain branching occurs. Linear polyethylene of types I and II is approximately 50 percent crystalline and types III and IV can be as high as 85 percent crystalline or more. Generally, polyethylene made by the low pressure process with a density below 0.960 is made by polymerizing ethylene with a small amount of comonomer such as propylene, 1-butene and/or 1-hexene.

Catalysts and processes for producing polyethylene are broadly disclosed in U.S. patents 3,887,494; 3,900,451; 4,053,436; 4,151,122; 4,236,988; 4,294,724; 4,347,158; 4,392,990; 4,394,291; and 4,405,501 which are hereby incorporated by reference.

## Low Molecular Weight Polyethylene Resin

The low molecular weight polyethylene resin can be any new or recycled ethylene homopolymer or ethylene copolymer. If a comonomer is used, the comonomer generally is an alpha-olefin with about 3 to about 20 carbon atoms per molecule. Preferably, the comonomer has from about 3 to about 10 carbon atoms per molecule and most preferably, from 3 to 8 carbon atoms per molecule. These types of comonomers are preferred due to their cost, availability, and associated properties. Examples of alpha-olefins useful in this invention as comonomers are propylene, 1-butene, 3-methyl-1-butene, 1-pentene, 3-methyl-1-pentene, 4-methyl-1-pentene, 1-hexene, and 3-ethyl-1-hexene. The amount of comonomer will generally comprise less than about 10 weight percent of the low molecular weight polyethylene resin. Preferably, the low molecular weight polyethylene resin, when employing a comonomer, will comprise from about zero to 4 weight percent comonomer. Generally, the characteristics of the low molecular weight

polyethylene resin are those listed below in Table I.

Table I

5	Low Molecular Weight Polyethylene Resin Characteristics							
	Characteristic	Broad Range	Preferred Range	Most Preferred Range				
10	Density in g/cm <sup>3</sup> Melt Index in g/10 min. at FR-190/2.16 Heterogeneity Index (Mw/Mn) <sup>2</sup> Weight Percent of Total Polymer <sup>1</sup>	D ≥ 0.94 MI > 25 2 ≤ H. I. ≤ 12 10 ≤ W. P. ≤ 80	0.95 ≤ D ≤ 0.98 25 < M. I. ≤ 800 2 ≤ H.I. ≤ 10 15 ≤ W. P. ≤ 70	$0.96 \le D \le 0.98$ $30 \le M. I. \le 500$ $2 \le H. I. \le 6$ $20 \le W. P. \le 60$				

<sup>&</sup>lt;sup>1</sup>Total polymer weight = weight of low molecular weight resin + weight of high molecular weight resin.

The low molecular weight polyethylene resin generally has a very low environmental stress crack resistance (ESCR).

#### High Molecular Weight Polyethylene Resin

The high molecular weight polyethylene resin can be any new or recycled ethylene copolymer resin. The comonomer generally is an alpha-olefin with about 3 to about 20 carbon atoms per molecule. Preferably, the comonomer has from about 3 to about 10 carbon atoms per molecule and most preferably, from 3 to 8 carbon atoms per molecule. Examples of alpha-olefins useful in this invention as comonomers are propylene, 1-butene, 3-methyl-1-butene, 1-pentene, 3-methyl-1-pentene, 4-methyl-1-pentene, 1-hexene, and 3-ethyl-1-hexene. The amount of comonomer will generally comprise less than about 40 weight percent of the high molecular weight polyethylene resin. Preferably, the high molecular weight polyethylene copolymer will comprise from about 0 to about 30 weight percent incorporated comonomer, and most preferably will comprise from 0.01 to 15 weight percent comonomer. Generally, the properties of the high molecular weight polyethylene resin are those listed below in Table II.

Table II

High Molecular Weight Polyethylene Resin Characteristics					
Characteristic	Broad Range	Preferred Range	Most Preferred Range		
High Load Melt Index in g/10 min. at FR-190/21.60 Heterogeneity Index	2 ≦ H.L.M.I. ≦ 10 2 ≦ H. I. ≦ 10	2 ≦ H.L.M.I. ≦ 9	2 ≦ H. I. ≦ 8		

In another embodiment of this invention the density of the high molecular weight polyethylene resin is between 0.93 and 0.95 grams per cubic centimeter. The high molecular weight polyethylene resin generally has a very high environmental stress crack resistance. Usually the environmental stress crack resistance of these polymers are so high that they cannot be practically measured.

#### **Blend Characteristics**

35

40

45

In general, the properties of the resulting blend of the low molecular weight polyethylene component and the high molecular weight polyethylene component are given in Table III below.

<sup>&</sup>lt;sup>2</sup>The Heterogeneity Index is an indication of the molecular weight distribution. This is a valuable indication of a resin's usefulness in certain applications. It is the weight average molecular weight (Mw) divided by the number average molecular weight (Mn).

#### Table III

Blend Characteristics							
Characteristic	Broad Range	Preferred Range	Most Preferred Range				
Density in g/cm <sup>3</sup>	0.94 ≤ D ≤ 0.98	0.942 ≤ D ≤ 0.970	0.945 ≤ D ≤ 0.965				
Melt Index in g/10 min. at FR-190/2.16	M.I. > 0.05	0.1 ≦ M.I. ≦ 5	0.1 ≦ M.I. ≦ 2				
Heterogeneity Index	2 ≦ H.I. ≦ 20	3 ≦ H.I. ≦ 15	6 ≤ H.I. ≤ 12				
Haze Percent	15 ≦ H ≦ 42	18 ≦ H ≦ 40	20 ≤ H ≤ 38				

### Blending of Components

The blending of the components can be accomplished by any method known in the art which produces a homogeneous blend from two polymer resins. Standard polymer processing equipment can be used to make these blends such as, for example, single screw extruders, twin screw extruders, and continuous mixers. Additionally, during blending other components can be added to the blend. These components can be antioxidants, UV stabilizers, preservatives, and processing aids such as fluoroelastomers. In general, "fluoroelastomer" is defined in this specification as any elastomeric polymer containing fluorine. They can be homopolymers or they can be copolymers. Such polymers tend to be amorphous, thermally stable and noncombustible, they also tend to have low glass transition temperatures and are generally resistant to a chemical attack by solvents and chemicals.

### **EXAMPLES**

40

45

55

This example is provided to further assist a person skilled in the art with understanding this invention. The particular reactants, conditions, and the like, are intended to be generally illustrative of this invention and are not meant to be construed as unduly limiting the reasonable scope of this invention.

The following test methods were used to determine the properties of the polyethylene samples.

- (1) The density was determined in accordance with ASTMD 1505-85. This is a standard test method for determining the density of plastics. The density is reported in grams per cubic centimeter ( $D = g/cm^3$ ).
- (2) The flow rate was determined in accordance with ASTMD 1238-65T. This is a standard test method for determining the flow rate of a thermoplastic. Two different conditions were used to determine the flow rate. Condition 1 consisted of a temperature of 190°C and a total load of 2.16 kilograms (FR-190/2.16) which is also called the melt index. Condition 2 consisted of a temperature of 190°C and a total load weight of 21.60 kilograms (FR-190/21.60) which is also called the high load melt index. The flow rate is reported as the rate of extrusion in grams per 10 minutes (g/10 min.).
- (3) The tensile strength at break was determined in accordance with ASTMD 638-90. This is a standard test method for determining the tensile properties of plastics. The tensile strength at break is reported in megapascals (MPa).
- (4) The percent elongation at break was determined in accordance with ASTMD 638-90. This is a standard test method for determining the tensile properties of plastics.
- (5) The Spencer impact was determined in accordance with ASTMD 3420-85. This is a standard test method for determining the pendulum impact resistance of plastic film. The Spencer impact is reported in Joules (J).
- (6) The dart impact was determined in accordance with ASTMD 1709-85. This is a standard test method for determining impact resistance of polyethylene film by the free falling dart method. The dart impact is reported in grams.
- (7) The tear resistance was determined in accordance with ASTMD 1922-89. This is a standard test method for determining the propagation tear resistance of plastic film and thin sheeting by the pendulum method. The tear resistance is reported in grams.
- (8) The environmental stress crack resistance (ESCR) was determined in accordance with ASTMD 1693-70. This is a standard test method for determining the environmental stress cracking resistance of ethylene plastics. The test specimens were prepared according to ASTMD 1928-80 procedure C. The condition of the test was condition A. The ESCR is reported in hours.
- (9) The haze was determined by using a Gardner XL-211 hazeguard/hazemeter system. As stated before, the haze is the amount of transmitted light that in passing through the specimen deviates from

the incident beam by forward scattering. Usually, in commercial haze meters only light deviating more than about 2.5° from the transmitted beam direction is considered haze. This device uses a procedure similar to ASTMD 1003-61.

(10) The heterogeneity index (Mw/Mn), which is the weight average molecular weight (Mw) divided by the number average molecular weight (Mn), was determined using data collected by gel permeation chromatography. This gel permeation chromatography was accomplished with a Waters 150C chromatograph at 140 °C using 1,2,4-trichlorobenzene as a solvent. This is a standard test method for determining molecular weight averages and molecular weight distributions.

#### 10 Example I

Blends of high and low molecular weight polyethylenes were prepared to demonstrate the properties of the blends of this invention. For each blend, the desired amounts of each polyethylene fluff (total of 260 g of polyethylene) were dry blended in a plastic container. Each polyethylene fluff sample was then mixed with acetone (10 mL/100 g of polyethylene), butylated hydroxytoluene (BHT), dilauryl thiodipropionate (DLTDP), and calcium stearate. The levels of additives in the final blend were 0.05 weight percent BHT, 0.03 weight percent DLTDP, and 0.04 weight percent calcium stearate, with the percentages based on the total polyethylene weight.

After the blends of fluff, acetone, and additives had been mixed by hand in a plastic container, the acetone wRs allowed to evaporate in a hood and the blend was then dried in a vacuum oven at 70-80 °C and about 380 mm of mercury vacuum for several hours. The dried blends were then mixed in a Midget Banbury mixer at a temperature between 150 - 160 °C for approximately 5 minutes at a mixing speed of 130 rpm and any large pieces were then ground in a mill.

The three high molecular weight and two low molecular weight polyethylenes used to produce the blends are described in Table E-I-A.

Table E-I-A
Polyethylene Blend Resins

	High Molecular Weight Polymer Resin	High Load Melt Index g/10 min FR-190/21.60	Density g/cm <sup>3</sup>
	1	1.0	0.9324
	2	3.1	0.9366
٠	3	6.4	0.9378
	Low Molecular Weight Polymer Resin	Melt Index g/10 min FR-190/2.16	Density, g/cm <sup>3</sup>
	4	31	0.9676
	5	312	0.9720

Molecular weight information about several of the polymers used in this example is presented in Table E-I-AI. Where the molecular weights were not determined (Polymers 1, 3, and 5), the molecular weights of polymers with similar melt viscosities (Polymers 1A, 3A, and 10) are listed. Polymer 10 is from Example II.

55

Table E-I-A1

Polyethylene Molecular Weights

Polymer	High Load Melt Index g/10 min	Mw Daltons	Mn Daltons	HI
1A	1.0	315,000	60,000	5.3
2	3.1	208,000	46,900	4.4
- 3A	6.0	189,000	41,900	4.5
			,	
Polymer	MI, g/10 min	Mw Daltons	Mn Daltons	HI
4	31	42,700	10,700	4.0
10	297	22,530	6,570	3.43

Nine blends, designated B1 through B9, were prepared. Their components, weight ratios, and properties are listed in Table E-I-B.

<sup>a</sup> Polymer numbers bweight ratio. <sup>c</sup> Not determined.	Н9	В8	В7	В6	B5	В4	85	B2	B1	Blend		
<sup>a</sup> Polymer numbers from Table E-I-A. <sup>b</sup> Weight ratio. <sup>c</sup> Not determined.	3/5 , 65/35	2/5 , 65/35	1/5 , 65/35	3/4 , 75/25	3/4 , 65/35	•	•	1/4 , 60/40	1/4 , 50/50	Blend Components High MV/Low MW <sup>a</sup> , ratio		
	n		n	0.48	0.69	0.36	0.54	0.16	0.26	Melt Index 8/10 min FR-190/2.16	1-10	
	33	17	7.1	15	24	13	22	7.8	15	High Load Melt Index 8/10 min FR-190/21.60	Polyethylene Blends	
	n	C	n	31	35	36	41	49	58	HI HTHI		
A Management of the Control of the C	0.9529	0.9512	0.9487	0.9471	0.9505	0.9487	0.9524	0.9477	0.9519	Density, 8/cm <sup>3</sup>		
	c	n	n	154,000	145,000	166,000	159,000	218,000	186,000	M⊌ Daltons		
	n	ი	c ·	21,100	19,500	19,900	17,100	21,000	18,000	Mn Daltons		
	n	n	ဂ	7.3	7.4	4.8	9.3	10.4	10.4	Heterogeneity Index		

Test specimens were compression molded from the blends and evaluated for properties. The results, summarized in Table E-I-C show the blends exhibit a good balance of properties. Non-blended polyethylene resins with a density of about 0.95 g/cm³ would usually have much lower ESCR values. For example, a polyethylene resin with a density of about 0.952 g/cm and a melt index of about 0.35g/10 minutes has an environmental stress crack resistance of about 50 hours. This resin just described is available from the Phillips Petroleum Company as HHM 5202.

Table E-I-C

Physical Properties						
Blend	Density, g/cm <sup>3</sup>	Tensile Break, MPa	Elongation at Break, %	ESCR, hrs		
B1	0.9519	36.9	1019	>1000		
B2	0.9477	36.6	955	>1000		
В3	0.9524	34.3	1029	198		
B4	0.9487	37.1	1028	>1000		
B5	0.9505	34.4	1061	616		
В6	0.9471	36.5	1054	>1000		
B7	0.9487	36.2	934	>1000		
B8	0.9512	31.9	920	>1000		
B9	0.9529	29.9	. 937	451		

### Example II

10

15

25

Several more blends of high and low molecular weight polyethylenes were prepared to show that blown films prepared from the blends were clearer than films made from non-blended, commercial, polyethylene blown film resins.

The four high molecular weight and three low molecular weight components for the blends are described in Table E-II-A. Polymer 5 is the same polymer as in Example I.

Table E-II-A
Polyethylene Samples

High Molecular Weight Polymer Resin	High Load Melt Index g/10 min FR-190/21.60	Density, g/cm <sup>3</sup>
6	3.1	0.9451
7	2.9	0.9366
8 .	3.6	0.9474
9	3.0	0.9399
Low Molecular Weight Polymer Resin	Melt Index g/10 min FR-190/2.16	Density,
10	297	0.9727
5	312	0.9720
11	96	0.9714

Molecular weight information was obtained on only Polymers 10 and 11. This information is shown in Table E-II-Al along with data for a polymer, designated Polymer 7A, similar in melt viscosity to Polymer 7:

Table E-II-Al

Polyethylene Molecular Weights								
Polymer	High Load Melt Index g/10 min FR-190/21.60	Melt Index g/10 min FR-190/2.16	Mw Daltons	Mn Daltons	ні			
7A	2.7	a	248,000	34,900	7.1			
10	a	297	22,530	6,570	3.43			
11	a	96	26,500	7,400	3.6			

Blends were prepared by mixing the components in a double cone blender with several additives. All polymer blends contained 0.05 weight percent bis(2,4-di-tert-butylphenyl)pentaerythritol diphosphite (Ultranox 626), 0.03 weight percent DLTDP, and 0.05 weight percent FX 9613 fluoroelastomer from 3M<sup>TM</sup>. Blend B10 also contained 0.06 weight percent BHT and 0.05 zinc stearate. The other blends also contained 0.02 weight percent BHT and 0.07 weight percent tetrakis(methylene 3-(3,5-di-t-butyl-4-hydroxyphenyl)-propionate)methane (Irganox 1010).

These blends, designated B10 through B14, were extruded with melt temperatures of 230-250 °C into pellets. The resulting resins are described in Table E-II-B with their original components and weight ratios.

Table E-II-B

Polyethylene Blends						
Blends	Blend Components High MW/Low MW <sup>a</sup> , ratio <sup>b</sup>	Blend High Load Melt Index g/10 min FR-190/21.60	Blend Density g/cm <sup>3</sup>			
B10	6/10 , 66/34	15.5	0.9579			
B11	7/5 , 67/33	15.9	0.9585			
B12	8/5 , 60/40	32.0	0.9613			
B13	8/11,60/40	26.6	0.9603			
B14	9/5 , 67/33	17.0	С			

- a Polymer numbers from Tables E-I-A and E-II-A.
- b Weight ratio.
- c Not determined.

These blends were extruded into blown film about 0.0254 mm thick using a 38 mm single screw extruder with a 50.8 mm film die with a melt temperature of 220-240°C for evaluation. Two control groups of polyethylene blown film resins, designated C1 and C2, were included for comparison. Control group resins C1 had a density of about 0.935 g/cm³ and a HLMI of about 15g/10 min. Control group resins C2 had a density of about 0.947 g/cm³ and a HLMI of about 17 g/10 min. Two extruder speeds were used in the film extrusion. Additionally, some of the control resins had 500 part per million by weight of a fluorelastomer. (Viton A<sup>TM</sup>).

The blown film properties listed in Table E-II-C show that the invention blends B10 through B15 have low haze values compared with the two control films, while retaining good overall film properties and higher densities. With blend B14, an increase in extruder speed from 35 to 75 rpm gave an increase in haze and little change in tear resistance strength. Overall, the blends had a tear strength and haze value usually associated with lower densities, yet these blends had much higher densities. This combination of high tear strength, lower haze, and higher density was unexpected.

5

10

25

30

#### Table E-II-C

		Filt	n Properties		
Blend	Screw Speed, rpm	Haze, %	Dart Drop, g	Spencer Impact, J	Tear Resistance, g MD/TD, <sup>c</sup>
B10	75	30.4	166	0.44	24.8/304
B11ª	75	32.4	128	0.41	b
	75	37.6	b	b	b
B12ª	35	37.0	88	0.30	b
	35	36.1	b	b	b
B13ª	35	33.0	88	0.31	b
	35	31.2	b	b	b
B14	35	24.6	212	b	32.3/454
	75	31.7	151	b	42.0/331
C1 (Control)	35 <sup>d</sup>	64.9	b	b	b
	75	47.8	151	b	43.0/317
	75 <sup>d</sup>	64.4	b	b	b
C2 (Control)	35 <sup>d</sup> 75 75 75 75	66.2 58.2 b b 70.8	b 193 180 164 b	b 0.60 0.58 0.49 b	b 22.1/408 17/400 b

<sup>&</sup>lt;sup>a</sup> Variable thickness film.

## 5 Claims

40

45

- 1. A composition of matter comprising:
  - (a) 10 to 80 weight percent of a low molecular weight polyethylene resin having a density of at least 0.94 grams per milliliter, a melt index greater than 25 grams per 10 minutes, and a heterogeneity index between 2 and 12; and
  - (b) 20 to 90 weight percent of a high molecular weight ethylene copolymer having a density less than 0.955 grams per milliliter; a high load melt index between 2 and 10 grams per 10 minutes, and a heterogeneity index between 2 and 10;
  - wherein said composition of matter has a melt index greater than 0.05 grams per 10 minutes.
- 2. The composition Of matter of claim 1 comprising:
  - (a) 20-60 weight percent of a low molecular weight ethylene homopolymer having a density between 0.96 and 0.98 g/ml, a melt index between 30 and 500 g/10 min., and a heterogeneity index between 2 and 6; and
  - (b) 40-80 weight percent of a high molecular weight ethylene/hexene copolymer having a density between 0.915 and 0.955 g/ml, a high load melt index between 2 and 8 g/10 min., and a heterogeneity index between 2 and 8, in particular wherein said density of said high molecular weight ethylene/hexene copolymer is between 0.93 and 0.95 grams per milliliter.
- 3. The composition of matter according to claim 1 wherein said low molecular weight resin is a copolymer.

<sup>&</sup>lt;sup>b</sup> Not determined.

<sup>&</sup>lt;sup>c</sup> MD = Machine direction.

TD = Transverse direction.

<sup>&</sup>lt;sup>d</sup> Had fluoroelastomer

- 4. The composition of matter according to claim 3 wherein said copolymer comprises ethylene and an alpha-olefin selected from the group consisting of propylene, 1-butene, 3-methyl-1-butene, 1-pentene, 3-methyl-1-pentene, 4-methyl-1-pentene, 1-hexene, 3-ethyl-1-hexene or mixtures thereof.
- The composition of matter according to claim 3 or 4 wherein said copolymer has a comonomer content of less than 10 weight percent, the weight percent based on the entire weight of the low molecular weight copolymer resin.
- 6. The composition of matter according to claim 1 wherein said high molecular weight ethylene copolymer comprises ethylene and an alpha-olefin selected from the group consisting of propylene, 1-butene, 3methyl-1-butene, 1-pentene, 3-methyl-1-pentene, 4-methyl-1-pentene, 1-hexene, 3-ethyl-1-hexene or mixtures thereof.
- 7. The composition of matter according to claim 6 wherein said high molecular weight ethylene copolymer has a comonomer content of less than 40 weight percent, the weight percent based on the total weight 15 of the high molecular weight ethylene copolymer resin.
  - The film made with a composition of matter of one of the preceding claims.
- The bottle made with a composition of matter of one of the preceding claims.

25

30

35

40

45

50

55



# **EUROPEAN SEARCH REPORT**

Application Number

EP 92 11 5931

ategory	Citation of document with it of relevant pa	ndication, where appropriate,	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
A		ITOMO CHEMICAL COMPANY	, 1-9	C08L23/04
				·
		•		
		• • • • • • • • • • • • • • • • • • • •		TECHNICAL FIELDS SEARCHED (Int. Cl.5)
				C08L
			÷	
			_	
	The present search report has b			Promises
	Place of search THE HAGUE	Date of completion of the search 25 NOVEMBER 1992		CLEMENTE GARCIA R.
X: pau Y: pau	CATEGORY OF CITED DOCUME rticularly relevant if taken alone rticularly relevant if combined with an cument of the same category	NTS T: theory or prin E: earlier patent after the fillin ther D: document cite	ciple underlying the document, but put g date d in the application d for other reasons	e invention blished on, or

THIS PAGE BLANK (USPTO)